

REPORT DOCUMENTATION PAGE

AFRL-SR-BL-TR-99-

0013

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1. AGENCY USE ONLY (Leave Blank)		2. REPORT DATE 19 January 1999	3. REPORT TYPE AND DATES COVERED Final Technical Report 09 Aug 96 - 09 Aug 98
4. TITLE AND SUBTITLE Materials for the "Plastic" Retina: Network Blends for Arrays of Polymer Grid Triodes Final Technical Report		5. FUNDING NUMBERS Contract# F49620-96-C-0041 <i>2303/cS</i> <i>61102F</i>	
6. AUTHOR(S) Dr. Jon S. McElvain		8. PERFORMING ORGANIZATION REPORT NUMBER ORTD4	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) UNIAX Corporation 6780 Cortona Drive Santa Barbara CA 93117		9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) Air Force Office of Scientific Research 110 duncan Avenue Room B115 Bolling AFB DC 20332-8080	
10. SPONSORING / MONITORING AGENCY REPORT NUMBER FA9550		11. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by the documentation.	
12 a. DISTRIBUTION / AVAILABILITY STATEMENT Approved for public release; distribution unlimited.		12 b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) We fabricated one-dimensional Plastic Retina structures by depositing gold strips onto glass substrates, and subsequently applying thin layers of conducting materials onto this pattern. The materials that were studied consisted of both conducting polymers (which were spin cast) and inorganic semiconductors. In all cases the thin film structures yielded results that were in accordance with theoretical predictions. The test structures were used to quantify the material sheet resistance uniformity and long-term stability. Additionally, we have synthesized several varieties of conducting polymer systems, including new PANI derivatives and blends. The ultimate goal of this effort was to create conducting polymer systems that have lower conductivity and higher film uniformity. This class of materials is essential to the operation of the Plastic Retina structures (currently being fabricated by UNIAX under a joint BMDO/DARPA program). For the Plastic Retina, or the Thin Film Analog Image Processor (TAIP), a sheet resistance greater than $10^5 \Omega/\text{square}$ is required. Furthermore, the TAIP requires a highly uniform (5%) PANI thickness across the surface of the chip. UNIAX has achieved this goal, and successfully fabricated new polymer systems that provide lower conductivities with high film uniformity.			
14. SUBJECT TERMS Smart Materials. Image Processing. Polyaniline. Conducting Polymers.			15. NUMBER OF PAGES 14
			16. PRICE CODE
17. SECURITY CLASSIFICATION OR REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION ON THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UL

NSN 7540-01-280-5500

Standard Form 298 (Rev.2-89)

Prescribed by ANSI Std. Z39-18

298-102

Materials for the “Plastic Retina”: Network Blends for Arrays of Polymer Grid Triodes

**Final Technical Report
January, 1999**

Contract number: F49620-96-C-0041

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I. Objectives

Since the device structure of the "Plastic Retina" has changed significantly, we have established new objectives for this project. The first objective is to demonstrate the use of a variety of thin film conducting materials for this new versatile image processing device. The goal here is to apply these materials to a one-dimensional test structure of the new "Plastic Retina" and test their operation characteristics, such as uniformity of resistivity and shelf life. The effort should reveal the best candidate materials for the two-dimensional Plastic Retina structure currently being fabricated under a joint BMDO/DARPA SBIR Phase II project. The second objective of this contract is synthesis of new forms of conducting polymers (such as polyaniline, PANI) which exhibit different conductivity ranges and greater film uniformity.

With regard to the first objective, this effort has been completed to our satisfaction. We fabricated one-dimensional Plastic Retina structures by depositing gold strips onto glass substrates, and subsequently applying thin layers of conducting materials onto this pattern. The materials that were studied consisted of both conducting polymers (which were spin cast) and inorganic semiconductors. Among the conducting polymers, the following systems were investigated: polyaniline (PANI), both in *m*-cresol and H₂O, polyethylene dioxythiophene-polystyrene sulfonate (PEDT-PSS) in H₂O, and blends of PANI-CSA with polyester (PES). Additionally, indium-tin-oxide (ITO) and doped amorphous silicon (a-Si), both inorganic semiconductors, were also used for these studies. In all cases the thin film structures yielded results that were in accordance with theoretical predictions. The test structures were used to quantify the material sheet resistance uniformity and long-term stability.

Regarding the second objective, we have synthesized several varieties of conducting polymer systems, including new PANI derivatives and blends. The ultimate goal of this effort was to create conducting polymer systems that have lower conductivity and higher film uniformity. This class of materials is essential to the operation of the Plastic Retina structures (currently being fabricated by UNIAX under a joint BMDO/DARPA program). For the Plastic Retina, or the Thin Film Analog Image Processor (TAIP), a sheet

resistance greater than $10^5 \Omega/\text{square}$ is required. Furthermore, the TAIP requires a highly uniform (5%) PANI thickness across the surface of the chip. UNIAX has successfully fabricated new polymer systems that provide lower conductivities with high film uniformity.

II. Accomplishments

A. Device Overview

In the initial phase of the Plastic Retina development, the device structure consisted of a two-dimensional array of Polymer Grid Triodes (Figure 1), whereby the grid was common to each of the pixels. The common grid provided lateral current flow between pixels and resulted in a form of “local contrast control”, similar to that implemented in the human retina.

The basic structure of the individual PGT is sketched in Figure 1; it is a complex multilayer structure consisting of various types of materials. The top layer (5) is the anode, the bottom layer (1) is the cathode. The third electrode (3), is the open network of PANI protonated to the highly conducting form with camphor sulfonic acid (CSA). Semiconducting polymer is used for layers (2) and (4), between the cathode and the polymer grid and between the PANI grid and the anode. *Because of the different work function of the cathode/anode (e.g. Ca or Al) and the PANI grid, layers (3),(4) and (5) and layers (1), (2) and (3) each form a diode in which the charge injection is limited by Fowler-Nordheim tunneling; denoted as a FN-diode.*

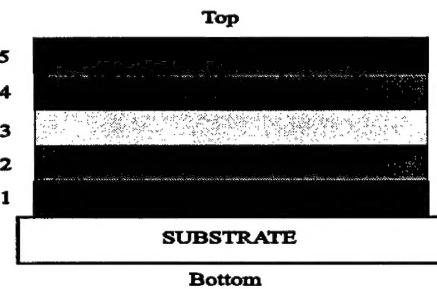


Fig 1: Structure of the polymer grid triode with the various layers. (1) and (5) are the cathode and anode (pixel) arrays, respectively. The other layers are continuous films common to all the PGTs within the array: (4) is the semiconducting polymer MEH PPV; (2) is the PVK resistor layer and (3) is the PANI common grid network filled with the semiconductor (3').

As a result of the research conducted during the AFOSR program, we discovered a significant simplification of the device structure for the Plastic Retina. Instead of having two semiconductor regions separated by a porous PANI network, the new structure consists of a single conducting polymer (or inorganic semiconductor) layer, interfaced with a group of identical capacitors. The architecture of the “new” Plastic Retina (TAIP) is shown in Figure 2; it consists of a simple layer of a conducting polymer coupled to a set of identical capacitors. This simple device functions as an active resistor-capacitor network. The charges on each capacitor are blurred by the lateral conduction afforded by the resistive polymer layer, resulting in an output which can be tuned to perform as either a low pass or a high pass *spatial* frequency filter.

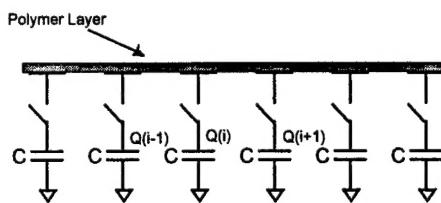


Figure 2: Schematic of the TAIP.

Initially, with all switches open, a charge $Q_0(i)$ is loaded onto each capacitor, where the charge in each unit cell represents a pixel value of the original (raw) image. At time $t=0$, all of the switches are closed simultaneously, and the charges $Q_0(i)$ are allowed to flow into the polymer film at a rate corresponding to the RC time constant of the circuit (where R is the polymer film resistance). Evidently, after the switches are closed, the charge on each capacitor will become a local average of its neighbors, i.e. $Q(i) = Q(i,t) = \langle Q(i) \rangle_t$. Obviously, after an infinite amount of time, all of these charges will be the same; in this case the number of pixels that contribute to the blurring average (the “blurring length”) will be equal to the array size. However, if the switches are opened after some time t_0 , the charge on each capacitor will be the local average of the surrounding pixels, with the number of pixels in the average determined by t_0 (relative to the RC time constant of the circuit).

The operation of this new TAIP (in one dimension) can be described by the following difference equation, which is obtained using Kirchoff's laws:

$$-RC \frac{dQ(i,t)}{dt} = 2Q(i,t) - Q(i+1,t) - Q(i-1,t) \quad (1)$$

where R is the pixel to pixel resistance of the polymer film, C is the unit cell capacitance, and Q(i) is the charge on capacitor i at time t .

Equation 1 is simply a discrete version of the one-dimensional diffusion equation; it can be easily generalized for two dimensions. For regions far from the array edges, the solution can be approximated by the following sum,

$$Q(i,t) = \langle Q_o \rangle_i \approx \frac{1}{2\sqrt{\pi}N_{eff}} \sum_j Q_o(j) \exp\left(-\frac{(i-j)^2}{4N_{eff}^2}\right) \quad (2)$$

N_{eff} is the decay parameter ("blurring length") of the device, and is defined as

$$N_{eff} = \sqrt{\frac{t}{RC}} \quad (3)$$

It describes the number of pixels over which the Gaussian blurring kernel of Equation 2 extends. Notice that the blurring length changes with time; as the switches remain closed, the number of pixels over which the averaging occurs becomes larger. This is consistent with intuition, as one expects a larger degree of charge averaging with increasing time.

Performing a Fourier transform of Equation 1, the modulation transfer function (MTF) for the grid voltage can be calculated (in k-space):

$$-RC \frac{dQ(k,t)}{dt} = 2Q(k,t)[1 - \cos(kc)] \quad (4)$$

where c is the spacing of the pixels in the array. The solution to Equation 3 is a simple exponential decay function, given by

$$Q(k,t) = Q_o(k) \exp\left\{-\frac{t}{RC} 2[1 - \cos(kc)]\right\} \quad (5)$$

where $\mathcal{Q}_o(k)$ is the Fourier transform of the initial ($t = 0$) charge distribution $\mathcal{Q}_o(i)$ (i.e. the Fourier transform of the raw image). Dividing Equation 5 by $\mathcal{Q}_o(k)$ yields the modulation transfer function (MTF) for the system,

$$MTF_{lp}(k) = \exp\left\{-\frac{t}{RC} 2[1 - \cos(kc)]\right\} \quad (6)$$

The MTF is essentially the transmission function in frequency space for the TAIP. The time dependent charge profile of the TAIP then behaves as a *low pass spatial frequency filter*. As demonstrated in Figure 3, spatial frequencies below $1/N_{eff}$ are passed, while those above this cutoff frequency are suppressed. Furthermore, since t is adjustable, this cutoff frequency is tunable; a large blurring length yields a low rolloff frequency, whereas a small blurring length yields a higher rolloff frequency.

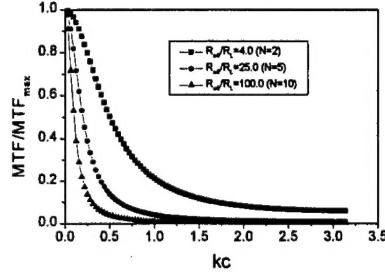


Figure 3: MTF curves for the TAIP low pass operation, for various blurring lengths. From Equations 3 and 6, by changing the time the switches remain closed, the frequency rolloff can be changed.

If the difference between the input voltage and the grid voltage is taken,

$$\Delta Q(i, t) = \mathcal{Q}_o(i) - \langle \mathcal{Q}_o(i) \rangle_t \equiv \mathcal{Q}_o(i) - \mathcal{Q}(i, t) \quad (7)$$

the following MTF for a *high pass spatial frequency filter* is generated:

$$MTF_{hp}(k) = 1 - \exp\left\{-\frac{t}{RC} 2[1 - \cos(kc)]\right\} \quad (8)$$

Equation 8 is plotted in Figure 4; it is evident that indeed a high pass filter is obtained, as the MTF curves all vanish at $k = 0$, and approach unity at higher spatial frequencies.

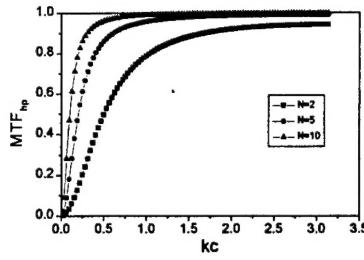


Figure 4: Demonstration of the MTF curves with the high pass filter implementation of the TAIP.

B. Device Fabrication and Characterization

Because of the potential utility of the TAIP, UNIAX has developed several one-dimensional (1x20) prototypes for its predecessor (Figure 5), with a variety of material bases. The device shown in Figure 5 functions in a similar fashion to the TAIP shown in Figure 2, with the capacitors replaced by variable resistors. In this case, the blur parameter is given by,

$$N_{eff} = \sqrt{\frac{R_v}{R_L}} \quad (9)$$

where R_v and R_L refer to the vertical resistance and the lateral polymer resistance, respectively. An “image” is applied to the input nodes of the device, and the output nodes (V_g) are monitored.

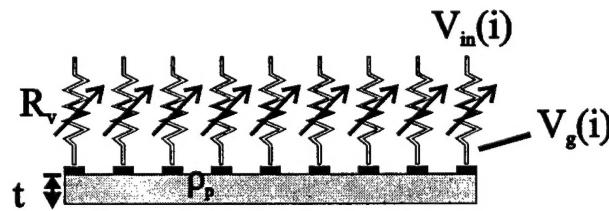


Figure 5: Schematic of the Plastic Retina devices characterized for basic image processing.

Shown below in Figure 6 is the grid voltage response to a step function input, for a device fabricated with the conducting form of polyaniline (PANI) spin coated onto patterned gold electrodes. For low values of R_v , the blurring length was very short, whereas for larger values of R_v the blurring length increased, consistent with Equation 9.

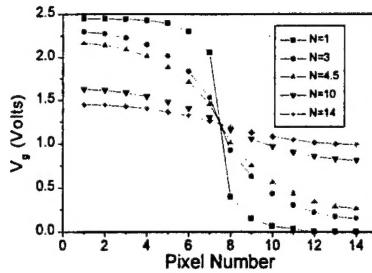


Figure 6: Step function response of a 1-D prototype Plastic Retina fabricated with conducting polyaniline (PANI), for different blurring lengths.

The sheet resistance of these films was also very uniform, as demonstrated in Figure 7, which shows the measured resistance between pixel 1 and pixel i . Clearly the data nearly follow a straight line, as expected for a film with high sheet resistance uniformity. Furthermore, these devices demonstrated high environmental stability, with the resistance of the PANI layer changing by less than 5% over a period of 2 months.

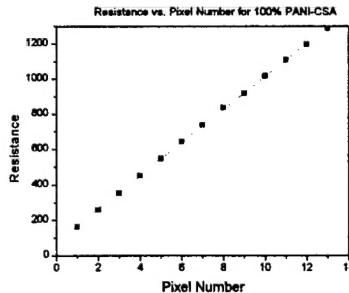


Figure 7: Resistance as a function of pixel number, for a device constructed with 100% PANI-CSA.

Similar devices have also been fabricated with several other conducting polymers, including PEDT and blends of PANI with PES, in addition to continuous films of indium tin oxide (ITO) and doped amorphous silicon. The fact that reliable devices have been fabricated with such a large variety of materials indicates the Plastic Retina has great promise for scale up to large format arrays. UNIAX has completed the fabrication of 320x256 TAIP arrays, as part of a joint BMDO/DARPA SBIR Phase II project, in an attempt to evaluate the utility of these devices for real imaging applications. Several of these TAIP devices are also being combined by UNIAX as part of a larger DARPA Smart Materials program to form versatile multipole bandpass spatial frequency filtering in analog hardware.

Because these structures are essentially resistive grids, they exhibit a great degree of fault tolerance; i.e. when one pixel is defective, it does not destroy the functionality of the entire device. Figure 8 demonstrates this fault tolerance quite clearly. The middle curve (squares) represents the response of an array to a step function (similar to that shown in Fig. 6). If the middle resistor is then removed from structure, the response curve is nearly identical to the defect-free case (figure 8, bottom curve). Furthermore, if the same resistor is replaced by another ten times smaller, the qualitative shape of the curve is unchanged (figure 8, top). This is an important feature when considering the production of large format arrays, since device operation must continue, even with a small number of dead pixels. This fault tolerance also applies to the capacitive approach sketched in Figure 2.

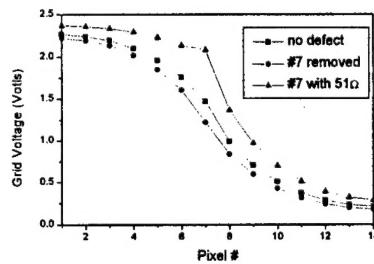


Figure 8: Demonstration of the fault tolerance of the Plastic Retina.

C. High Resistance PANI Compound Synthesis

Template-guided synthesis of conducting polymers was first reported by S.C. Yang [L. Sun, S.C.Yang, Polymer Reprints, **33**, 379 (1992)]. The molecular template, in most cases, polyacids such as polystyrene sulfonic acid, binds the monomer, for example aniline, to form molecular complexes. The attractive force between template and monomer can be chemical and/or physical. Upon polymerization, the aniline monomers form polyaniline and remain attached to the template to form the template-polyaniline complex. By judicious choice of the template molecule and the polymerization conditions, stable sub-micron size colloidal particles of polyaniline-template aggregate can be formed during polymerization. The colloid particles are stabilized by the surface charge provided by extra sulfonic acid group in polystyrene sulfonic acid. Yang succeeded in the synthesis of a polyaniline-polystyrenesulfonic acid complex in an

aqueous medium. The synthesized complex can be dispersed very well in water to form a stable dispersion with particle size less than 1 μm . Transparent films with specific resistivity 1-10 $\Omega\text{-cm}$ can then be cast from such dispersions.

Subsequent work used a similar approach to prepare aqueous dispersions of polyaniline (PANI), the conductivity of which can be controlled over a wide range. Films prepared from such dispersions are used as the hole-injection layer for polymer LED displays, and can be used for image processing applications such as the TAIP. Aqueous polyaniline dispersions were also developed independently [M. Angelopoulos, N. Patil, J. Shaw, Mat. Res. Soc. Symp., 328, 173 (1994); M. Angelopoulos, J.D. Gelorme, T.H. Newman, N. M. Patil, D.E. Seeger, US patent, 5,370,825] for use as removable charge dissipaters in the e-beam patterning process.

Recently, Bayer [G. Heywang, F. Jonas, Adv. Mater., 4, 116 (1992); J. Friedrich, K. Werner, B. Aktiengesellschaft, US patent, 5,300,575] commercialized poly(ethylenedioxythiophene)-polystyrene sulfonate (PEDT-PSS), under the trade name TP AI4071. The PEDT-PSS is prepared as a stable dispersion in water. Films of PEDT-PSS are semi-transparent; they can be spin-cast with a surface resistance of approximately 500 Ω/square and with 75% transmission. The ITO/PEDT-PSS bilayer has also been used as the anode in LEDs with external quantum efficiency ca. 2% and with good stress life [Y. Cao, G. Yu, C. Zhang, R. Menon, A.J. Heeger, Synth. Met., 87, 171 (1997)].

Despite the obvious advantage of the conducting polymers in aqueous dispersions mentioned above, a major disadvantage for these materials is their high conductivity. For the TAIP application, this conductivity is over 2 orders of magnitude too high, as a sheet resistances in excess $10^5 \Omega/\text{square}$ is required. The goal of this research effort was therefore to produce new PANI compounds that produced highly uniform films with relatively high resistances. We have approached this problem in two ways: 1) looking for new polymer protonic acids and 2) by blending the resulting PANI with commercially available host polymers. The procedures and results are discussed below.

For the first stage, PANI-PAAMPSA was prepared according ref. [Y.Cao, et al, Polymer, **30** (1989) 2307]. For this procedure, HCl from the reference was replaced by poly(2-acrylamido-2-methyl-1-propanesulfonic acid (PAAMPSA) (Aldrich). 30.5 g (0.022 mole) of 15% PAAMPSA in water (Aldrich) was deluted to 2.3% by adding 170 ml water. While stirring, 2.2 g (0.022M) aniline was added into the PAAMPSA solution. 2.01 g (.0088M) of Ammonium persulfate in 10 ml water was added slowly into the aniline/PAAMPSA solution under vigorous stirring. The reaction mixture was stirred for 24 hours at room temperature. 1000 ml acetone was added into reaction mixture to precipitate the product (PANI-PAAMPSA). Most of the acetone/water was decanted and then the PANI-PAAMPSA precipitate was filtrated. The resulting gum-like product was washed several times with acetone and was dried at 40°C under dynamic vacuum for 24 hours. The product was then redissolved in DI water and re-precipitated by acetone. Finally, a 1-4% (w/w) solution of final product in water was prepared by vigorous stirring of PANI-PAAMPSA powder in DI water at RT for 24 hours in plastic container. The solution was filtrated through 0.45 μ m filter before using.

For the second stage, the aqueous solution of PANI-PAAMPSA was mixed with an aqueous solution of polyacrylamide (PAcAm). These PANI blend films were then spin-coated on top of glass substrates deposited with Au electrodes. The resistance between the Au electrodes was measured using a Keithley high resistance meter. In order to calculate the sheet resistance, the resulting film thickness was measured using a DEKTAC thickness profiler.

Table 1 shows the measured conductivity of the polyblend films with different blend compositions. The second column refers to the relative ratio (w/w) of the two host polymers in the blend. It is clear that the resulting conductivity can be varied over four orders of magnitude, simply by changing the blend ratio. Even more important to the TAIP architecture is the fact that the *sheet* resistance (resistivity/thickness) can be changed over a much broader range, since it is trivial to increase or decrease the thickness of the PANI film.

As a comparison, we list in Table 2 conductivity and sheet resistance values for commercially available PEDT (Bayer) polyblend solutions currently being evaluated at UNIAX for our LED display applications. Again in this case, conductivity can be controlled over three orders of magnitude, with a larger mean conductivity of approximately 10^4 S/cm.

ID	Ratio of host polymers*	Thickness (Å)	ohm/sq	σ (S/cm)
	<u>PAAMPSA/PAcAm</u>			
3-19-3	2/0	2080	8.5×10^8	5.6×10^{-5}
		1300	2.1×10^9	3.7×10^{-5}
3-28-1	0.5/1	1850	6.4×10^{10}	9.3×10^{-7}
		1000	3.6×10^{11}	2.8×10^{-7}
3/24	1/1	1620	5.9×10^{10}	1.0×10^{-6}
		1100	1.4×10^{12}	6.5×10^{-8}
3-26-2	0/2	1200	1.0×10^{12}	8.3×10^{-8}
		750	1.8×10^{13}	7.4×10^{-9}

* Ratio of polyaniline to the individual host polymers is 1/2(w/w)

Table 1: Measured conductivity and sheet resistance for various blend ratios of PANI-PAAMPSA and PANI-PAcAm.

Type	Batch	Spin speed (RPM)	Thickness (Å)	Rs (MΩ/sq)	σ (S/cm)
PEDT-PSS	33/97	600	1000	8.5	1.2×10^{-2}
		1000	760	10.1	1.3×10^{-2}
PANI blend	4-7-1'	1000	2100	522	9.0×10^{-5}
		2000	1500	1550	4.3×10^{-5}
		3000	1200	4480	1.9×10^{-5}
		4000	1000	5300	1.9×10^{-5}

Table 2. Thickness and conductivity of new PEDT-PSS in comparison with PANI blend.

The wide range of conductivity control for conducting polymers is essential for flexible operation of the TAIP chip. For the TAIP architecture being fabricated under our existing BMDO/DARPA Phase II SBIR, the blur time is controllable over 4 orders of magnitude, from 130 nanoseconds to 1.3 milliseconds. Referring to Equation 3, if one wishes to achieve a blurring parameter range of 1 to 100, the RC time constant must be set to 130 nanoseconds. Assuming a unit cell capacitance of 300 femtoFarads, this implies $R=433\text{k}\Omega$. However, if instead it is desired to achieve a blurring parameter ranging from 0.1 to 10, the pixel-to pixel resistance required is $R=43\text{M}\Omega$. It is therefore clear that the ability to choose from a wide range of conducting polymers is essential to maximizing the flexibility of the TAIP architecture. This has been successfully demonstrated during the course of this contract.

III. Personnel Supported

Dr. Jon McElvain has carried out the bulk of the research related to this project, specifically focusing on the device fabrication. Dr. Yong Cao contributed to the PANI process development, along with Mark Rafetto. Prof. Alan Heeger was also partially supported under this contract, serving primarily as technical supervisor.

IV. Publications

During the first term of this contract, we submitted one paper, appearing in the International Conference on Image Processing (ICIP'97) proceedings. The publication is entitled "Spatial Frequency Filtering Using Non-Delineated Thin Films". Additionally, UNIAX will be submitting a paper to the SPIE 6th Annual International Symposium on Smart Structures and Materials, entitled "Spatial Frequency Filtering Using Hybrid Polymer/VLSI Technology". This paper/presentation will summarize to date the Plastic Retina program at UNIAX.

V. Interactions/Transitions

UNIAX Corp. attended the ICIP'97, held in Santa Barbara, CA Oct. 26-29, 1997. An oral presentation was given on Oct. 29 regarding the technology developed during this

contract. A paper also appeared in the proceedings of this conference summarizing our progress.

The research performed under this contract has had a direct impact on a large Smart Materials program recently established by DARPA. A component of this program will involve combining several of these Plastic Retina devices to create a super-architecture capable of complex image processing functions. The ultimate goal of this system is to enhance target detectability while providing a low power, high speed, and compact implementation. This technology will be of particular interest to target track/detection applications, for example cruise missile detection systems, where complex image processing functions must be performed in real time in order to reduce false alarm rates. The individuals sponsoring this program at DARPA are Dr. Steve, Dr. Dennis Healy, Dr. Frank Patten, and Dr. Randy Sands. Dr Joel Davis at NRL will serve as the executing agent for the UNIAX portion of the program. Dr. Dean Scribner at NRL will be collaborating with UNIAX on the test and measurement of the system. Additionally, Raytheon Infrared Center of Excellence (RIRCOE, formerly SBRC) will be participating in the device/system design and development throughout the course of the program.

VI. New Discoveries

As a result of the research performed during the course of this contract, significant modifications to the original Plastic Retina device structure have occurred. Consequently, we have filed a provisional patent entitled "Thin Film Spatial Filters" (application # 60/053572), which details the various image processing capabilities of this device.